

TRANSMITTAL LETTER TO THE UNITED STATES

ATTORNEY'S DOCKET NUMBER 49651

DESIGNATED/ELECTED OFFICE (DO/EO/US)
 CONCERNING A FILING UNDER 35 U.S.C. 371

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

INTERNATIONAL APPLICATION NO.
 PCT/EP 99/09803

INTERNATIONAL FILING DATE
 11 December 1999

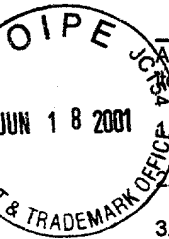
PRIORITY DATE CLAIMED
 19 December 1998

TITLE OF INVENTION: FUNGICIDAL MIXTURES BASED ON MORPHOLINE OR PIPERDINE DERIVATIVES AND
 OXIME ETHER DERIVATIVES

APPLICANT(S) FOR DO/EO/US Klaus SCHELBERGER, Maria SCHERER, Reinhold SAUR, Karl EICKEN, Egon HADEN,
 Eberhard AMMERMAN, Thomas GORTE, Gisela LORENZ, Siegfried STRATHMANN

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following
 items and other information:

1. ☒ This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
 2. ☐ This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
 3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
 4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
 5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/USO).
 6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
 7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).
 - a. ☒ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
 8. ☒ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
 9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 171(c)(4)).
 10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).
- Items 11. to 16. below concern other document(s) or information included:
11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
 12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
 13. ☒ A FIRST preliminary amendment.
☐ A SECOND or SUBSEQUENT preliminary amendment.
 14. ☐ A substitute specification.
 15. ☐ A change of power of attorney and/or address letter.
 16. ☒ Other items or information.
 International Search Report
 International Preliminary Examination Report



RECEIVED
 JUN 18 2001
 U.S. PATENT & TRADEMARK OFFICE

U.S. Appln. No. (If Known) INTERNATIONAL APPLN. NO.
PCT/EP99/09803

ATTORNEY'S DOCKET NO.
49651

		CALCULATIONS	PTO USE ONLY
17. /X/ The following fees are submitted			
BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):			
Search Report has been prepared by the EPO or JPO.....	\$860.00	860.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482).....	\$750.00		
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)).....	\$700.00		
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO	\$ 970.00		
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied pro-visions of PCT Article 33(2)-(4).....	\$96.00		
ENTER APPROPRIATE BASIC FEE AMOUNT = \$		860.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than / / 20 / / 30 months from the earliest claimed priority date (37 CFR 1.492(e)).			
<u>Claims</u>	<u>Number Filed</u>	<u>Number Extra</u>	<u>Rate</u>
Total Claims	10 -20		X\$18.
Indep. Claims	1 -3		X\$80.
Multiple dependent claim(s) (if applicable)	+270.		
TOTAL OF ABOVE CALCULATION		=	860.00
Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28).			
SUBTOTAL		=	860.00
Processing fee of \$130. for furnishing the English translation later than / / 20 / / 30 months from the earliest claimed priority date (37 CFR 1.492(f)).			
TOTAL NATIONAL FEE		=	860.00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) \$40.00 per property			
TOTAL FEES ENCLOSED		= \$	900.00
		Amount to be refunded: \$	
		Charged \$	

a./X/ A check in the amount of \$ 900. to cover the above fees is enclosed.

b./ / Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c./X/ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 11-0345. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:
KEIL & WEINKAUF
1101 Connecticut Ave., N.W.
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SIGNATURE

Herbert B. Keil
NAME

Registration No. 18,967

09/868515

JC18 Rec'd PCT/PTO 1 8 JUN 2001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of)
SCHELBERGER et al.) BOX PCT

International Application)
PCT/EP 99/09803)

Filed: December 11, 1999)

For: FUNGICIDAL MIXTURES BASED ON MORPHOLINE OR PIPERIDINE
DERIVATIVES AND OXIME ETHER DERIVATIVES

PRELIMINARY AMENDMENT

Honorable Commissioner of
Patents and Trademarks
Washington, D.C. 20231

Sir:

Prior to examination, kindly amend the above-identified application as follows:

IN THE CLAIMS

Please amend the claims as shown in the attached sheet.


REMARKS

The claims were amended in the preliminary examination. The claims have been amended further to eliminate multiple dependency and to put them in better form for U.S. filing. No new matter is included. A clean copy of the claims is attached.

Favorable action is solicited.

Respectfully submitted,

KEIL & WEINKAUF


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(202)659-0100

CLEAN VERSION OF THE AMENDED CLAIMS - OZ 49651

4. A fungicidal mixture as claimed in claim 1, where in the compounds II, R³ or R⁴ are hydrogen, fluorine, chlorine, methyl, ethyl, methoxy, thiomethyl or N-methyamino.

9. A fungicidal mixture as claimed in claim 1, which is conditioned in two parts, where one part comprises one or more compounds I in a solid or liquid carrier and the other part comprises one or more compounds of the formula II in a solid or liquid carrier.

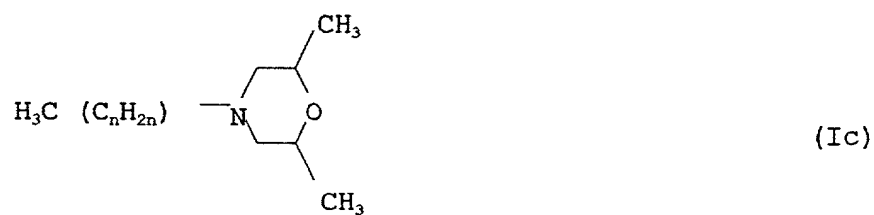
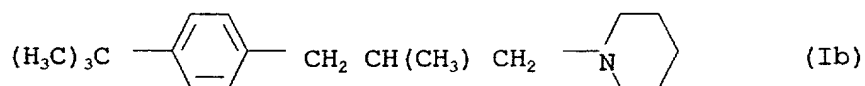
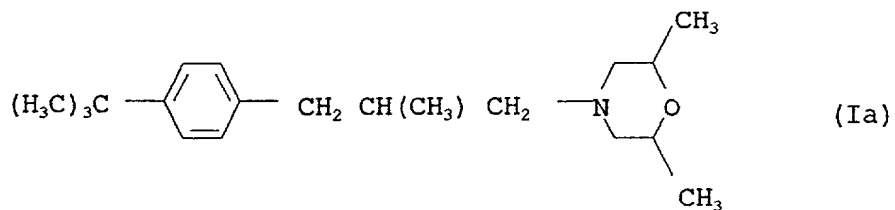
10. A method for controlling harmful fungi, which comprises treating the fungi, their habitat or the materials, plants, seeds, soils, areas or spaces to be protected against fungal attack with a fungicidal mixture as claimed in claim 1, where the compounds I and one or more compounds of the formulae II can be applied simultaneously, that is either together or separately, or successively.

MARKED UP VERSION OF THE CLAIMS - OZ 49651

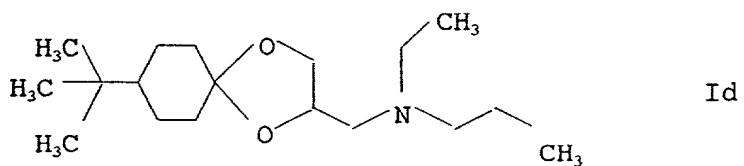
4. A fungicidal mixture as claimed in claim 1, where in the compounds II, R³ or R⁴ are hydrogen, fluorine, chlorine, methyl, ethyl, methoxy, thiomethyl or N-methy-amino [[sic]].
9. A fungicidal mixture as claimed in claim 1 [any one of the preceding claims], which is conditioned in two parts, where one part comprises one or more compounds I in a solid or liquid carrier and the other part comprises one or more compounds of the formula II in a solid or liquid carrier.
10. A method for controlling harmful fungi, which comprises treating the fungi, their habitat or the materials, plants, seeds, soils, areas or spaces to be protected against fungal attack with a fungicidal mixture as claimed in claim 1 [any of claims 1 to 9], where the compounds I and one or more compounds of the formulae [[sic]] II can be applied simultaneously, that is either together or separately, or successively.

CURRENT CLAIMS - OZ 49651

1. A fungicidal mixture, comprising as active components
 - a) a morpholine or piperidine derivative I selected from the group of the compounds Ia, Ib, Ic and Id

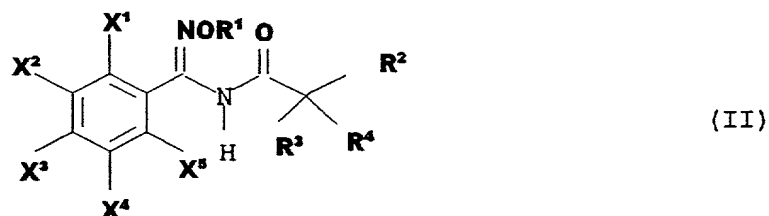


[n= 10,11,12 (60 - 70%) or 13]



and

b) compounds of the formula II



where the substituents X^1 to X^5 and R^1 to R^4 are as defined below:

X^1 is C_1 - C_4 -haloalkyl, C_1 - C_4 -haloalkoxy or halogen;

X^2 to X^5 are, independently of one another, hydrogen, halogen, C_1 - C_4 -alkyl, C_1 - C_4 -haloalkyl, C_1 - C_4 -alkoxy or C_1 - C_4 -haloalkoxy,

R^1 is C_1 - C_4 -alkyl, C_2 - C_6 -alkenyl, C_2 - C_6 -alkynyl, C_1 - C_4 -alkyl- C_3 - C_7 -cycloalkyl, where these radicals may carry substituents selected from the group consisting of halogen, cyano and C_1 - C_4 -alkoxy,

R^2 is a phenyl radical or a 5- or 6-membered saturated or unsaturated

heterocyclyl radical having at least one heteroatom selected from the group consisting of N, O and S, where the cyclic radicals may have one to three substituents selected from the group consisting of halogen, C₁-C₄-alkyl, C₁-C₄-alkoxy, C₁-C₄-haloalkyl, C₁-C₄-haloalkoxy, C₁-C₄-alkoxy-C₂-C₄-alkenyl, C₁-C₄-alkoxy-C₂-C₄-alkynyl,

R³ and R⁴ are, independently of one another, hydrogen, C₁-C₄-alkyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, N-C₁-C₄-alkylamino, C₁-C₄-haloalkyl or C₁-C₄-haloalkoxy

in a synergistically effective amount.

2. A fungicidal mixture as claimed in claim 1, where in the compounds II, R¹ is C₁-C₄-alkyl or C₁-C₄-alkylene-C₃-C₇-cycloalkyl.
3. A fungicidal mixture as claimed in claim 1, where in the compounds II, R² is phenyl, thienyl, pyrazolyl, pyrrolyl, imidazolyl, thiazolyl, furyl, pyridazinyl or pyrimidinyl, and these radicals may be substituted by halogen, C₁-C₄-alkoxy or C₁-C₄-alkyl.
4. A fungicidal mixture as claimed in claim 1, where in the compounds II, R³ or R⁴ are

hydrogen, fluorine, chlorine, methyl, ethyl, methoxy, thiomethyl or N-methymino.

5. A fungicidal mixture as claimed in claim 1, where in the compounds II, X^1 is halo- C_1-C_6 -alkyl, halo- C_1-C_6 -alkoxy or halogen.
6. A fungicidal mixture as claimed in claim 1, where in the compounds II, X^2 or X^3 are hydrogen or halogen.
7. A fungicidal mixture as claimed in claim 1, where in the compounds II, X^4 is hydrogen, chlorine, fluorine, methoxy, ethoxy, trifluoromethyl or trifluoromethoxy.
8. A fungicidal mixture as claimed in claim 1, where in the compounds II, X^5 is hydrogen, chlorine, fluorine, methoxy, ethoxy, trifluoromethyl or trifluoromethoxy.
9. A fungicidal mixture as claimed in claim 1, which is conditioned in two parts, where one part comprises one or more compounds I in a solid or liquid carrier and the other part comprises one or more compounds of the formula II in a solid or liquid carrier.
10. A method for controlling harmful fungi, which comprises treating the fungi, their habitat or the materials, plants, seeds, soils, areas or spaces to be protected

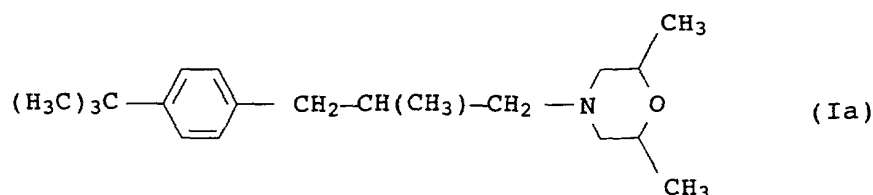
against fungal attack with a fungicidal mixture as claimed in claim 1, where the compounds I and one or more compounds of the formulae II can be applied simultaneously, that is either together or separately, or successively.

Fungicidal mixtures based on morpholine or piperidine derivatives
and oxime ether derivatives

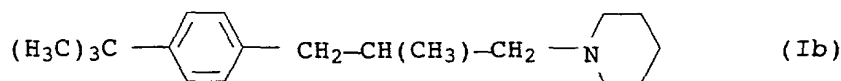
5 The present invention relates to fungicidal mixtures for controlling harmful fungi and to methods for controlling harmful fungi using such mixtures.

WO 97/40673 provides fungicidal mixtures which, inter alia,
10 comprise active compounds of the formulae Ia, Ib and/or Ic in addition to other fungicidally active compounds from the group of the oxime ethers and/or the carbamates.

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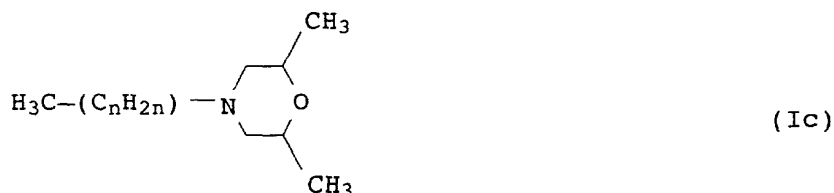


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[n = 10, 11, 12 (60 - 70%) or 13]

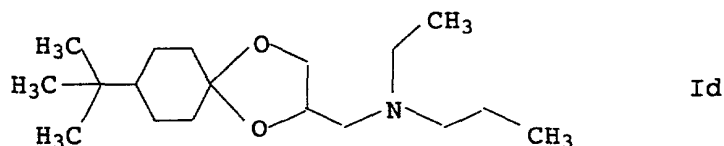
Other fungicidal mixtures which comprise active compounds of the formulae Ia to Ic are disclosed in EP-A 797386, WO 97/06681, EP-B 425857, EP-B 524496, EP-A 690792, WO 94/22308 and EP-B 645087.

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Brighton Crop Protection Conference 1996, Pests and Diseases, pp.47-52 discloses the active compound of the formula Id:

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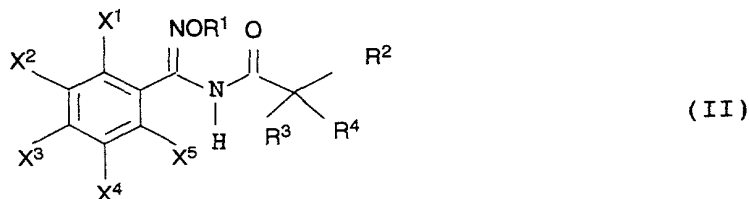
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DE 19722223 describes mixtures of compounds of the formula II and of active compounds from the class of the strobilurins.

It is an object of the present invention to provide other particularly effective mixtures for controlling harmful fungi and, in particular, for certain indications.

We have found that this object is surprisingly achieved with a mixture which, as active compounds, comprises morpholine or piperidine derivatives of the formula I defined at the outset and, as further fungicidally active component, at least one fungicidally active compound of the formula II

20



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where the substituents X^1 to X^5 and R^1 to R^4 are as defined below:

X^1 is C_1 - C_4 -haloalkyl, C_1 - C_4 -haloalkoxy or halogen

30

X^2 to X^5 are, independently of one another, hydrogen, halogen, C_1 - C_4 -alkyl, C_1 - C_4 -haloalkyl, C_1 - C_4 -alkoxy or C_1 - C_4 -haloalkoxy;

35

R^1 is C_1 - C_4 -alkyl, C_2 - C_6 -alkenyl, C_2 - C_6 -alkynyl, C_1 - C_4 -alkyl- C_3 - C_7 -cycloalkyl, where these radicals may carry substituents selected from the group consisting of halogen, cyano and C_1 - C_4 -alkoxy,

40

R^2 is a phenyl radical or a 5- or 6-membered saturated or unsaturated heterocyclyl radical having at least one heteroatom selected from the group consisting of N, O and S, where the cyclic radicals may have one to three substituents selected from the group consisting of halogen, C_1 - C_4 -alkyl, C_1 - C_4 -alkoxy, C_1 - C_4 -haloalkyl, C_1 - C_4 -haloalkoxy, C_1 - C_4 -alkoxy- C_2 - C_4 -alkenyl, C_1 - C_4 -alkoxy- C_2 - C_4 -alkynyl,

45

3

R^3 and R^4 are, independently of one another, hydrogen,
 C_1 - C_4 -alkyl, C_1 - C_4 -alkoxy, C_1 - C_4 -alkylthio,
 N - C_1 - C_4 -alkylamino, C_1 - C_4 -haloalkyl or
 C_1 - C_4 -haloalkoxy

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in a synergistically effective amount.

The mixtures according to the invention act synergistically, and
 they are therefore particularly suitable for controlling harmful
 10 fungi and, in particular, powdery mildew fungi.

In the context of the present invention, halogen is fluorine,
 chlorine, bromine and iodine and in particular fluorine, chlorine
 and bromine.

15

The term "alkyl" includes straight-chain and branched alkyl
 groups. These are preferably straight-chain or branched
 C_1 - C_{12} -alkyl groups and in particular C_1 - C_6 -alkyl groups. Examples
 of alkyl groups are alkyl, such as, in particular, methyl, ethyl,
 20 propyl, 1-methylethyl, butyl, 1-methylpropyl, 2-methylpropyl
 [lacuna] 1,1-dimethylethyl, n-pentyl, 1-methylbutyl,
 2-methylbutyl, 3-methylbutyl, 1,2-dimethylpropyl,
 1,1-dimethylpropyl, 2,2-dimethylpropyl, 1-ethylpropyl, n-hexyl,
 1-methylpentyl, 2-methylpentyl, 3-methylpentyl, 4-methylpentyl,
 25 1,2-dimethylbutyl, 1,3-dimethylbutyl, 2,3-dimethylbutyl,
 1,1-dimethylbutyl, 2,2-dimethylbutyl, 3,3-dimethylbutyl,
 1,1,2-trimethylpropyl, 1,2,2-trimethylpropyl, 1-ethylbutyl,
 2-ethylbutyl, 1-ethyl-2-methylpropyl, n-heptyl, 1-methylhexyl,
 1-ethylpentyl, 2-ethylpentyl, 1-propylbutyl, octyl, decyl,
 30 dodecyl.

Haloalkyl is an alkyl group which is defined as above and is
 partially or fully halogenated by one or more halogen atoms, in
 particular by fluorine and chlorine. Preferably, there are 1 to 3
 35 halogen atoms present, and particular preference is given to the
 difluoromethane [sic] and the trifluoromethyl group.

The alkenyl group includes straight-chain and branched
 C_2 - C_6 -alkenyl groups. Examples of alkenyl groups are 2-propenyl,
 40 2-butenyl, 3-butenyl, 1-methyl-2-propenyl, 2-methyl-2-propenyl,
 2-pentenyl, 3-pentenyl, 4-pentenyl, 1-methyl-2-butenyl,
 2-methyl-2-butenyl, 3-methyl-2-butenyl, 1-methyl-3-butenyl,
 2-methyl-3-butenyl, 3-methyl-3-butenyl, 1,1-dimethyl-2-propenyl,
 1,2-dimethyl-2-propenyl, 1-ethyl-2-propenyl, 2-hexenyl,
 45 3-hexenyl, 4-hexenyl, 5-hexenyl, 1-methyl-2-pentenyl,
 2-methyl-2-pentenyl, 3-methyl-2-pentenyl, 4-methyl-2-pentenyl,
 1-methyl-3-pentenyl, 2-methyl-3-pentenyl, 3-methyl-3-pentenyl,

- 4-methyl-3-pentenyl, 1-methyl-4-pentenyl, 2-methyl-4-pentenyl, 3-methyl-4-pentenyl, 4-methyl-4-pentenyl, 1,1-dimethyl-2-butenyl, 1,1-dimethyl-3-butenyl, 1,1-dimethyl-3-butenyl, 1,2-dimethyl-2-butenyl, 1,2-dimethyl-3-butenyl,
- 5 1,3-dimethyl-2-butenyl, 1,3-dimethyl-3-butenyl, 2,2-dimethyl-3-butenyl, 2,3-dimethyl-2-butenyl, 2,3-dimethyl-3-butenyl, 1-ethyl-2-butenyl, 1-ethyl-3-butenyl, 2-ethyl-2-butenyl, 2-ethyl-3-butenyl, 1,1,2-trimethyl-2-propenyl, 1-ethyl-1-methyl-2-propenyl and 1-ethyl-2-methyl-2-propenyl, in
- 10 particular 2-propenyl, 2-butenyl, 3-methyl-2-butenyl and 3-methyl-2-pentenyl.

The alkenyl group may be partially or fully halogenated by one or more halogen atoms, in particular by fluorine and chlorine. It

15 has preferably 1 to 3 halogen atoms.

- The alkynyl group includes straight-chain and branched C₃-C₆-alkynyl groups. Examples of alkynyl groups are 2-propynyl, 2-butyne, 3-butyne, 1-methyl-2-propynyl, 2-pentyne,
- 20 3-pentyne, 4-pentyne, 1-methyl-3-butyne, 2-methyl-3-butyne, 1-methyl-2-butyne, 1,1-dimethyl-2-propynyl, 1-ethyl-2-propynyl, 2-hexynyl, 3-hexynyl, 4-alkynyl, 5-hexynyl, 1-methyl-2-pentyne, 1-methyl-3-pentyne, 1-methyl-4-pentyne, 2-methyl-3-pentyne, 2-methyl-4-pentyne, 3-methyl-4-pentyne, 4-methyl-2-pentyne,
- 25 1,2-dimethyl-2-butyne, 1,1-dimethyl-3-butyne, 1,2-dimethyl-3-butyne, 2,2-dimethyl-3-butyne, 1-ethyl-2-butyne, 1-ethyl-3-butyne, 2-ethyl-3-butyne and 1-ethyl-1-methyl-2-propynyl.

- 30 The C₁-C₄-alkylene-C₃-C₇-cycloalkyl group is a C₃-C₇-cycloalkyl group, such as cyclopropyl, cyclobutyl, cyclopentyl or cyclohexyl, which is attached via a C₁-C₄-alkylene radical.

- Suitable substituents R² are, in addition to phenyl (unsubstituted
- 35 or substituted), in particular thienyl, pyrazolyl, pyrrolyl, imidazolyl, thiazolyl, furyl, pyridazinyl and pyrimidinyl. Preferred substituents at these ring systems are halogen (in particular F and Cl), C₁-C₄-alkoxy (in particular methoxy) and C₁-C₄-alkyl (in particular methyl, ethyl). The number of the ring
- 40 substituents can be from 1 to 3 and is in particular 1 or 2. Particular preference is given to phenyl or substituted phenyl, thienyl, thienyl-C₁-C₄-alkyl, pyrazolyl and pyrazol-C₁-C₄-alkyl.

- The substituents R³ and R⁴ are C₁-C₄-alkyl, C₁-C₄-alkoxy,
- 45 C₁-C₄-alkylthio, N-C₁-C₄-alkylamino, C₁-C₄-haloalkyl or C₁-C₄-haloalkoxy. Preferred substituents R³ and R⁴ are hydrogen,

F, Cl, methyl, ethyl, methoxy, thiomethyl and N-methylamino. R^3 and R^4 together may also form a grouping =O.

The morpholine or piperidine derivatives I (Ia: common name:

- 5 Fenpropimorph, US-A 4,202,894; Ib: common name: Fenpropidin, US-A 4,202,894; Ic: common name: Tridemorph, DE-A 11 64 152), their preparation and their action against harmful fungi are known, and they are commercially available products.

- 10 The compounds of the formula II and processes for their preparation are described in WO-A 96/19442 and in the earlier applications DE 1 97 41098.7 and 1 97 41099.5.

Among the compounds of the formula II, preference is given to

- 15 those where X^1 is a C_1 - C_4 -haloalkyl, in particular a trifluoromethyl group, a C_1 - C_4 -haloalkoxy, in particular a difluoromethoxy or trifluoromethoxy group or a halogen, in particular chlorine and X^2 and X^3 are a hydrogen atom or a halogen group, in particular a hydrogen atom. X^4 and X^5 are preferably
- 20 hydrogen, halogen (in particular Cl or F), C_1 - C_4 -alkoxy (in particular methoxy or ethoxy), C_1 - C_4 -haloalkyl (in particular trifluoromethyl) or C_1 - C_4 -haloalkoxy (in particular trifluoromethoxy).
- 25 Preferred substituents R^1 are C_1 - C_4 -alkyl (methyl, ethyl, n- and isopropyl and t-butyl), C_1 - C_4 -alkylene- C_3 - C_7 -cycloalkyl, C_1 - C_4 -alkenyl (in particular ethenyl, propenyl and butenyl, which may be substituted, in particular by halogen (preferably Cl)), propynyl, cyanomethyl and methoxymethyl. Among the
- 30 C_1 - C_4 -alkylene- C_3 - C_7 -cycloalkyl substituents, methylene-substituted compounds, in particular methylenecyclopropyl, methylenecyclopentyl, methylenecyclohexyl and methylenecyclohexenyl, are particularly preferred. The rings in these substituents may be substituted, preferably by halogen.
- 35 Suitable substituents R^2 are, in addition to phenyl (unsubstituted or substituted), in particular thienyl, pyrazolyl, pyrrolyl, imidazolyl, thiazolyl, furyl, pyridazinyl and pyrimidinyl. Preferred substituents at these ring systems are halogen (in
- 40 particular F and Cl), C_1 - C_4 -alkoxy (in particular methoxy) and C_1 - C_4 -alkyl (in particular methyl, ethyl). The number of the ring substituents can be from 1 to 3 and is in particular 1 or 2. Particular preference is given to phenyl or substituted phenyl.

Preferred compounds of the formula II are shown in the tables of WO 96/019442, which has already been mentioned. Among these compounds, in turn, particular preference is given to the compounds listed in Table 1 below (R^3 and R^4 are each hydrogen).

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Table 1:

	No.	X ¹	X ²	X ³	X ⁴	X ⁵	R ¹	R ²
10	II.1	CF ₃	H	H	H	H	ethyl	Ph-4-OMe
	II.2	CF ₃	H	H	H	H	methyl	Ph-4-OMe
	II.3	CF ₃	H	H	H	H	-CH ₂ -cPr	2-thienyl
	II.4	CF ₃	H	H	H	H	-CH ₂ -cPr	3-thienyl
15	II.5	CF ₃	H	H	H	H	-CH ₂ -cPr	Ph-2,4-F ₂
	II.6	CF ₃	H	H	H	H	-CH ₂ -cPr	Ph-2-F
	II.7	CF ₃	H	H	H	H	-CH ₂ -cPr	Ph-2-F-4-OMe
	II.8	CF ₃	H	H	H	H	-CH ₂ -cPr	Ph-3-Me
20	II.9	CF ₃	H	H	H	H	-CH ₂ -cPr	Ph-3-Me-4-OMe
	II.10	CF ₃	H	H	H	H	-CH ₂ -cPr	Ph-4-F
	II.11	CF ₃	H	H	H	H	-CH ₂ -cPr	Ph-4-Me
	II.12	CF ₃	H	H	H	H	-CH ₂ -cPr	Ph-4-OMe
25	II.13	CF ₃	H	H	H	H	-CH ₂ -cPr	Ph
	II.14	CF ₃	H	H	H	H	-CH ₂ -CH=CH ₂	Ph
	II.15	CF ₃	H	H	H	H	-CH ₂ -CH=CH ₂	Ph-4-OMe
	II.16	CF ₃	H	H	H	H	-CH ₂ -CH=CCl ₂	Ph-4-OMe
30	II.17	CF ₃	H	H	H	F	-CH ₂ -CH ₃	Ph-4-OMe
	II.18	CF ₃	H	H	H	F	-CH ₂ CH ₃	Ph
	II.19	CF ₃	H	H	H	F	-CH ₃	Ph-4-OMe
	II.20	CF ₃	H	H	H	F	-CH ₂ -cPr	Ph
35	II.21	CF ₃	H	H	H	F	-CH ₂ -cPr	Ph-2-F
	II.22	CF ₃	H	H	H	F	-CH ₂ -cPr	Ph-2,4-F ₂
	II.23	CF ₃	H	H	H	F	-CH ₂ -cPr	Ph-2-F-3-Me
	II.24	CF ₃	H	H	H	F	-CH ₂ -cPr	Ph-2-F-4-OMe
40	II.25	CF ₃	H	H	H	F	-CH ₂ -cPr	Ph-3,5-Me ₂
	II.26	CF ₃	H	H	H	F	-CH ₂ -cPr	3-methylpyrazol-1-yl
	II.27	CF ₃	H	H	H	F	-CH ₂ -cPr	3-methyl-2-thienyl
	II.28	CF ₃	H	H	H	F	-CH ₂ -cPr	2-thienyl
45	II.29	CF ₃	H	H	H	F	-CH ₂ -cPr	3-thienyl
	II.30	CF ₃	H	H	H	F	-CH ₂ -CHF ₂	Ph-4-OMe
	II.31	CF ₃	H	H	H	F	-CH ₂ -OCH ₃	Ph-4-OMe
	II.32	CF ₃	H	H	H	F	-CH ₂ -OCH ₃	Ph

No.	X ¹	X ²	X ³	X ⁴	X ⁵	R ¹	R ²	
5	II.33	CF ₃	H	H	H	F	-CH ₂ CN	Ph-4-Ome
	II.34	CF ₃	H	H	H	F	-CH ₂ CN	Ph
	II.35	CF ₃	H	H	H	F	-CH ₂ -C≡CH	Ph
	II.36	CF ₃	H	H	H	F	-CH ₂ -C≡CH	Ph-4-OMe
	II.37	CF ₃	H	H	H	F	-CH ₂ -C≡CH	Ph-2-F
10	II.38	CF ₃	H	H	H	F	-CH ₂ -C≡CH	Ph-4-Me
	II.39	CF ₃	H	H	H	F	-CH ₂ -C≡CH	2-thienyl
	II.40	CF ₃	H	H	H	F	-CH ₂ -C≡CH	Ph-2-F-4-OMe
	II.41	CF ₃	H	H	H	F	i-propyl	Ph
	II.42	CF ₃	H	H	H	F	n-butyl	Ph
15	II.43	CF ₃	H	H	H	F	n-propyl	Ph
	II.44	CF ₃	H	H	H	F	t-butyl	Ph
	II.45	CF ₃	H	H	H	Cl	-CH ₃	
	II.46	CF ₃	H	H	H	Cl	-CH ₂ CN	Ph-4-OMe
	II.47	CF ₃	H	H	H	Cl	-CH ₂ -OMe	Ph-4-OMe
20	II.48	CF ₃	H	H	H	Cl	-CH ₂ -cPr	Ph
	II.49	CF ₃	H	H	H	Cl	-CH ₂ -cPr	3-methylpyra- zol-1-yl
	II.50	CF ₃	H	H	H	Cl	-CH ₂ -cPr	2-thienyl
	II.51	CF ₃	H	H	H	Cl	-CH ₂ -cPr	Ph-2,4-F ₂
	II.52	CF ₃	H	H	H	Cl	-CH ₂ -C≡CH	Ph-4-OMe
25	II.53	CF ₃	H	H	H	CF ₃	-CH ₃	Ph-4-OMe
	II.54	CF ₃	H	H	H	CF ₃	-CH ₂ CH ₂ Cl	Ph-4-OMe
	II.55	CF ₃	H	H	H	CF ₃	-CH ₂ -cPr	2-thienyl
	II.56	CF ₃	H	H	H	CF ₃	-CH ₂ -cPr	Ph-2-F-5-Me
	II.57	CF ₃	H	H	H	CF ₃	-CH ₂ -cPr	Ph-4-OMe
30	II.58	CF ₃	H	H	H	CF ₃	-CH ₂ -cPr	Ph
	II.59	CF ₃	H	H	H	OCH ₃	-CH ₂ CH ₃	Ph-4-OMe
	II.60	CF ₃	H	H	H	OCH ₃	-CH ₂ -cPr	Ph-4-OMe
	II.61	CF ₃	H	H	H	OCH ₃	-CH ₂ -cPr	Ph
	II.62	CF ₃	H	H	Cl	F	-CH ₂ -CH ₂ Cl	Ph
35	II.63	CF ₃	H	H	Cl	F	-CH ₂ -CH=CH ₂	Ph-4-OMe
	II.64	CF ₃	H	H	Cl	F	-CH ₂ -cPr	2-thienyl
	II.65	CF ₃	H	H	Cl	F	-CH ₂ -cPr	Ph-2-F
	II.66	CF ₃	H	H	Cl	F	-CH ₂ -cPr	Ph
	II.67	CF ₃	H	H	Cl	F	-CH ₂ -cPr	Ph-2-F-5-Me
40	II.68	CF ₃	H	H	Cl	Cl	-CH ₂ -CH=CH ₂	Ph-4-OMe
	II.69	CF ₃	H	H	Cl	Cl	-CH ₂ CH ₂ Cl	Ph
	II.70	CF ₃	H	H	Cl	Cl	-CH ₂ CH ₃	Ph-2-F-5-Me

No.	X ¹	X ²	X ³	X ⁴	X ⁵	R ¹	R ²	
5	II.71	CF ₃	H	H	Cl	Cl	-CH ₂ -cPr	Ph-3,5-Me ₂
	II.72	CF ₃	H	H	SCH ₃	F	-CH ₂ -cPr	Ph-4-OMe
	II.73	CF ₃	H	H	OCH ₃	F	-CH ₂ -cPr	Ph-4-OMe
	II.74	CF ₃	H	F	H	H	-CH ₂ -cPr	Ph
	II.75	CF ₃	H	F	H	H	-CH ₂ -CH ₃	Ph-4-OMe
10	II.76	CF ₃	H	H	F	F	-CH ₂ CH ₃	Ph
	II.77	CF ₃	H	H	F	F	-CH ₂ -CH ₂ Cl	Ph-2-F-5-Me
	II.78	CF ₃	H	H	F	F	-CH ₂ -OCH ₃	Ph-4-OMe
	II.79	CF ₃	H	H	F	F	-CH ₂ -cPr	Ph
	II.80	CF ₃	H	H	F	F	-CH ₂ -cPr	3-methylpyra- zol-1-yl
15	II.81	CF ₃	H	H	F	F	-CH ₂ -cPr	3-methyl- 2-thienyl
	II.82	CF ₃	H	H	F	F	-CH ₂ -cPr	Ph-2-F-3-Me
	II.83	CF ₃	H	H	F	F	-CH ₂ -cPr	Ph-2-F-4-OMe
	II.84	CF ₃	H	H	F	F	-CH ₂ -cPr	Ph-2-F-5-Me
	II.85	CF ₃	H	H	F	F	-CH ₂ -cPr	Ph-4-OMe
20	II.86	CF ₃	H	H	F	F	-CH ₂ -cPr	Ph-4F
	II.87	CF ₃	H	H	F	F	i-propyl	Ph-4-OMe
	II.88	CF ₃	H	H	F	F	n-butyl	Ph-4-OMe
	II.89	CF ₃	H	H	F	F	-CH ₂ -C≡CH	Ph-4-OMe
	II.90	CF ₃	H	H	CF ₃	F	-CH ₃	Ph-4-OMe
25	II.91	CF ₃	H	H	CF ₃	F	-CH ₂ -CH=CH ₂	Ph
	II.92	CF ₃	H	H	CF ₃	F	-CH ₂ -cPr	Ph
	II.93	CF ₃	H	H	Cl	Cl	-CH ₂ -cHxe-3	Ph
	II.94	CF ₃	H	H	F	H	-CH ₂ -cPr	Ph-4-F
	II.95	CF ₃	H	H	Cl	Cl	-CH ₂ -cHex	Ph
30	II.96	CF ₃	H	H	H	F	-CH ₂ -SCH ₃	Ph
	II.97	CF ₃	H	H	H	F	-CH ₂ -SOCH ₃	Ph
	II.98	CF ₃	H	H	H	F	-CH ₂ -SO ₂ CH ₃	Ph
	II.99	CF ₃	H	H	H	F	-CH ₂ -NHMe	Ph
	II.100	CF ₃	H	H	H	F	CH ₂ -CONH ₂	Ph
35	II.101	CF ₃	H	H	H	F	CH ₂ CON(CH ₃) ₂	Ph

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In the table above, cPr is cyclopropyl, cHxe-n is cyclohexenyl which is unsaturated in position n, c-Hex is cyclohexyl and Ph is phenyl.

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Particular preference is given to compounds II in which R^1 is a radical CH_2-cPr and R^2 is an unsubstituted or substituted phenyl radical. Among these, in turn, preference is given to the compounds in which X^4 and X^5 [sic] are halogen, preferably F.

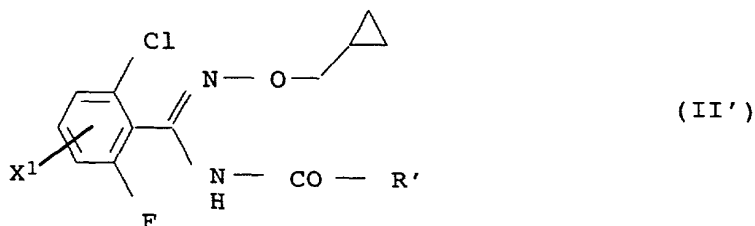
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Other preferred compounds of the formula II are shown in Tables 2 and 3 below.

Table 2: Compounds of the formula II'

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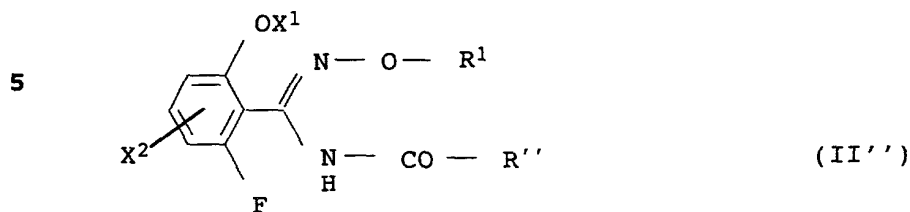
where the substituents are as defined below:

	No.	X ¹	R'	m.p. (°C)
25	II.102	H	4-CH ₃ -C ₆ H ₄ -CH ₂	86-88
	II.103	H	4-F-C ₆ H ₄ -CH ₂	79-81
	II.104	H	4-Cl-C ₆ H ₄ -CH ₂	105-107
	II.105	H	4-CH ₃ O-C ₆ H ₄ -CH ₂	73-76
30	II.106	H	4-CF ₃ -C ₆ H ₄ -CH ₂	
	II.107	5-F	4-CH ₃ -C ₆ H ₄ -CH ₂	87-90
	II.108	5-F	4-F-C ₆ H ₄ -CH ₂	71-74
	II.109	5-F	4-Cl-C ₆ H ₄ -CH ₂	85-87
35	II.110	5-F	4-CH ₃ O-C ₆ H ₄ -CH ₂	90-92
	II.111	5-F	4-CF ₃ -C ₆ H ₄ -CH ₂	
	II.112	H	2-thienylmethyl	87-89
	II.113	H	3-thienylmethyl	
40	II.114	5-F	2-thienylmethyl	90-93
	II.115	5-F	3-thienylmethyl	
	II.116	5-F	3-CH ₃ -C ₆ H ₄ -CH ₂	72-75
	II.117	5-F	2-F-C ₆ H ₄ -CH ₂	73-76
	II.118	5-F	4-CH ₂ FO-C ₆ H ₄ -CH ₂	oil

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Table 3: Compounds of the formula II''



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No.	OX ¹	X ²	R ¹	R''	m.p. °C
II.119	CHF ₂	H	C ₂ H ₅	C ₆ H ₅ -CH ₂	
II.120	CHF ₂	H	C ₂ H ₅	4-CH ₃ O-C ₆ H ₄ -CH ₂	
15 II.121	CHF ₂	H	CH ₂ -CH=CH ₂	C ₆ H ₅ -CH ₂	
II.122	CHF ₂	H	CH ₂ -C≡CH	C ₆ H ₅ -CH ₂	
II.123	CHF ₂	H	CH ₂ -C≡CH	4-CH ₃ O-C ₆ H ₄ -CH ₂	
II.124	CHF ₂	H	cPr	C ₆ H ₅ -CH ₂	
20 II.125	CF ₃	H	cPr	C ₆ H ₅ -CH ₂	
II.126	CHF ₂	H	cPr	4-F-C ₆ H ₄ -CH ₂	75-77
II.127	CHF ₂	H	cPr	4-Cl-C ₆ H ₄ -CH ₂	81-83
II.128	CHF ₂	H	cPr	4-CH ₃ O-C ₆ H ₄ -CH ₂	57-59
25 II.129	CHF ₂	H	cPr	4-CF ₃ -C ₆ H ₄ -CH ₂	
II.130	CHF ₂	H	cPr	2-thienylmethyl	oil
II.131	CHF ₂	H	cPr	3-thienylmethyl	oil
II.132	CHF ₂	H	cPr	pyrazolyl-1-methyl	
II.133	CHF ₂	H	cPr	4-CH ₃ -C ₆ H ₄ -CH ₂	
30 II.134	CHF ₂	5-F	CH ₂ -CH=CH ₂	C ₆ H ₅ -CH ₂	
II.135	CHF ₂	5-F	CH ₂ -CH=CH ₂	4-CH ₃ -C ₆ H ₄ -CH ₂	
II.136	CHF ₂	5-F	CH ₂ -C≡CH	C ₆ H ₅ -CH ₂	
II.137	CHF ₂	5-F	CH ₂ -C≡CH	4-CH ₃ O-C ₆ H ₄ -CH ₂	
35 II.138	CHF ₂	5-F	cPr	C ₆ H ₅ -CH ₂	62-65
II.139	CHF ₂	5-F	cPr	4-F-C ₆ H ₄ -CH ₂	64-67
II.140	CHF ₂	5-F	cPr	4-Cl-C ₆ H ₄ -CH ₂	72-75
II.141	CHF ₂	5-F	cPr	4-CH ₃ -C ₆ H ₄ -CH ₂	74-76
40 II.142	CHF ₂	5-F	cPr	4-CH ₃ O-C ₆ H ₄ -CH ₂	79-81
II.143	CHF ₂	5-F	cPr	4-CF ₃ -C ₆ H ₄ -CH ₂	
II.144	CF ₃	5-F	cPr	C ₆ H ₅ -CH ₂	
II.145	CHF ₂	4-F	cPr	C ₆ H ₅ -CH ₂	
II.146	CHF ₂	4-F	cPr	4-CH ₃ O-C ₆ H ₄ -CH ₂	
45 II.147	CHF ₂	H	cPr	4-CH ₃ -C ₆ H ₄ -CH ₂	69-71

The physical data of these compounds and processes for their preparation are given in the already mentioned WO 96/19442, DE 197441098.7 and DE 19741099.5.

- 5 The ratios of the compounds I and II can be varied within wide ranges; the active compounds are preferably employed in a ratio by weight in the range from 20:1 to 1:20, in particular 10:1 to 1:10.
- 10 When preparing the mixtures, it is preferred to employ the pure active ingredients I and II, to which further active ingredients against harmful fungi or other pests, such as insects, arachnids or nematodes, or else herbicidal or growth-regulating active ingredients or fertilizers can be admixed.
- 15 The mixtures of the compounds I and II, or the compounds I and II used simultaneously, jointly or separately, exhibit outstanding activity against a wide range of phytopathogenic fungi, in particular from the classes of the Ascomycetes, Basidiomycetes, 20 Phycomycetes and Deuteromycetes. Some of them act systemically and can therefore be employed as foliar- and soil-acting fungicides.
- They are especially important for controlling a large number of 25 fungi in a variety of crop plants, such as cotton, vegetable species (e.g. cucumbers, beans, tomatoes, potatoes and cucurbits), barley, grass, oats, bananas, coffee, maize, fruit species, rice, rye, soya, grapevine, wheat, ornamentals, sugar cane, and a variety of seeds.
- 30 They are particularly suitable for controlling the following phytopathogenic fungi: *Erysiphe graminis* (powdery mildew) in cereals, *Erysiphe cichoracearum* and *Sphaerotheca fuliginea* in cucurbits, *Podosphaera leucotricha* in apples, *Uncinula necator* in 35 grapevines, *Puccinia* species in cereals, *Rhizoctonia* species in cotton, rice and lawns, *Ustilago* species in cereals and sugar cane, *Venturia inaequalis* (scab) in apples, *Helminthosporium* species in cereals, *Septoria nodorum* in wheat, *Botrytis cinerea* (gray mold) in strawberries, vegetables, ornamentals and 40 grapevines, *Cercospora arachidicola* in groundnuts, *Pseudocercospora herpotrichoides* in wheat and barley, *Pyricularia oryzae* in rice, *Phytophthora infestans* in potatoes and tomatoes, *Plasmopara viticola* in grapevines, *Pseudoperonospora* species in hops and cucumbers, *Alternaria* 45 species in vegetables and fruit, *Mycosphaerella* species in bananas and *Fusarium* and *Verticillium* species.

They can furthermore be employed in the protection of materials (for example the protection of wood), for example against *Paecilomyces variotii*.

- 5 The compounds I and II can be applied simultaneously, that is either together or separately, or successively, the sequence, in the case of separate application, generally not having any effect on the result of the control measures.
- 10 Depending on the kind of effect desired, the application rates of the mixtures according to the invention are, in particular in agricultural crop areas, from 0.01 to 10 kg/ha, preferably 0.1 to 5 kg/ha, in particular 0.2 to 3.0 kg/ha.

- 15 The application rates of the compounds I are from 0.01 to 2.5 kg/ha, preferably 0.01 to 10 kg/ha, in particular 0.05 to 5.0 kg/ha.

- Correspondingly, in the case of the compounds II, the application rates are from 0.01 to 2 kg/ha, preferably 0.02 to 2 kg/ha, in particular 0.02 to 1.0 kg/ha.

- For seed treatment, the application rates of the mixture are generally from 0.001 to 250 g/kg of seed, preferably 0.01 to 25 100 g/kg, in particular 0.01 to 50 g/kg.

- If phytopathogenic harmful fungi are to be controlled, the separate or joint application of the compounds I and II or of the mixtures of the compounds I and II is effected by spraying or 30 dusting the seeds, the plants or the soils before or after sowing of the plants, or before or after plant emergence.

- The fungicidal synergistic mixtures according to the invention or the compounds I and II can be formulated for example in the form 35 of ready-to-spray solutions, powder and suspensions or in the form of highly concentrated aqueous, oily or other suspensions, dispersions, emulsions, oil dispersions, pastes, dusts, materials for broadcasting or granules, and applied by spraying, atomizing, dusting, broadcasting or watering. The use form depends on the 40 intended purpose; in any case, it should ensure as fine and uniform as possible a distribution of the mixture according to the invention.

- The formulations are prepared in a known manner, e.g. by 45 extending the active compound with solvents and/or carriers, if desired using emulsifiers and dispersants, it being possible also to use other organic solvents as auxiliary solvents if water is

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- used as the diluent. Suitable auxiliaries for this purpose are essentially: solvents such as aromatics (e.g. xylene), chlorinated aromatics (e.g. chlorobenzenes), paraffins (e.g. mineral oil fractions), alcohols (e.g. methanol, butanol),
- 5 ketones (e.g. cyclohexanone), amines (e.g. ethanolamine, dimethylformamide) and water; carriers such as ground natural minerals (e.g. kaolins, clays, talc, chalk) and ground synthetic minerals (e.g. finely divided silica, silicates); emulsifiers such as nonionic and anionic emulsifiers (e.g. polyoxyethylene
- 10 fatty alcohol ethers, alkylsulfonates and arylsulfonates) and dispersants such as lignosulfite waste liquors and methylcellulose.

- Suitable surfactants are the alkali metal salts, alkaline earth
- 15 metal salts and ammonium salts of aromatic sulfonic acids, e.g. ligno-, phenol-, naphthalene- and dibutyl-naphthalenesulfonic acid, and of fatty acids, alkyl- and alkylarylsulfonates, alkyl, lauryl ether and fatty alcohol sulfates, and salts of sulfated hexa-, hepta- and octadecanols, or of fatty alcohol glycol
- 20 ethers, condensates of sulfonated naphthalene and its derivatives with formaldehyde, condensates of naphthalene or of the naphthalenesulfonic acids with phenol and formaldehyde, polyoxyethylene octylphenol ether, ethoxylated isooctyl-, octyl- or nonylphenol, alkylphenol or tributylphenyl polyglycol ethers,
- 25 alkylaryl polyether alcohols, isotridecyl alcohol, fatty alcohol/ethylene oxide condensates, ethoxylated castor oil, polyoxyethylene alkyl ethers or polyoxypropylene, lauryl alcohol polyglycol ether acetate, sorbitol esters, lignosulfite waste liquors or methylcellulose.

30

Powders [lacuna] materials for broadcasting and dusts can be prepared by mixing or jointly grinding the compounds I or II or the mixture of the compounds I and II with a solid carrier.

- 35 Granules (e.g. coated granules, impregnated granules or homogeneous granules) are usually prepared by binding the active compound, or active compounds, to a solid carrier.

- Fillers or solid carriers are, for example, mineral earths, such
- 40 as silicas, silica gels, silicates, talc, kaolin, limestone, lime, chalk, bole, loess, clay, dolomite, diatomaceous earth, calcium sulfate, magnesium sulfate, magnesium oxide, ground synthetic materials and fertilizers, such as ammonium sulfate, ammonium phosphate, ammonium nitrate, ureas, and products of
- 45 vegetable origin, such as cereal meal, tree bark meal, wood meal and nutshell meal, cellulose powders or other solid carriers.

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The formulations generally comprise from 0.1 to 95% by weight, preferably 0.5 to 90% by weight, of one of the compounds I or II or of the mixture of the compounds I and II. The active compounds are employed in a purity of from 90% to 100%, preferably 95% to 5 100% (according to NMR spectrum or HPLC spectrum [sic]).

The compounds I or II, the mixtures, or the corresponding formulations, are applied by treating the harmful fungi, their habitat, or the plants, seeds, soils, areas, materials or spaces 10 to be kept free from them with a fungicidally effective amount of the mixture, or of the compounds I and II in the case of separate application.

15 Application can be effected before or after infection by the harmful fungi.

Examples of such preparations comprising the active compounds are:

- 20 I. A solution of 90 parts by weight of the active compounds and 10 parts by weight of N-methylpyrrolidone; this solution is suitable for use in the form of microdrops;
- II. A mixture of 20 parts by weight of the active compounds, 80 25 parts by weight of xylene, 10 parts by weight of the adduct of 8 to 10 mol of ethylene oxide to 1 mol of oleic acid N-monoethanolamide, 5 parts by weight of the calcium salt of dodecylbenzenesulfonic acid, 5 parts by weight of the adduct of 40 mol of ethylene oxide to 1 mol of castor oil; 30 a dispersion is obtained by finely distributing the solution in water;
- III. An aqueous dispersion of 20 parts by weight of the active compounds, 40 parts by weight of cyclohexanone, 30 parts by weight of isobutanol, 20 parts by weight of the adduct of 40 mol of ethylene oxide to 1 mol of castor oil;
- 35 IV. An aqueous dispersion of 20 parts by weight of the active compounds, 25 parts by weight of cyclohexanol, 65 parts by weight of a mineral oil fraction of boiling point 210 to 280°C, and 10 parts by weight of the adduct of 40 mol of ethylene oxide to 1 mol of castor oil;
- 40 V. A mixture, ground in a hammer mill, of 80 parts by weight of the active compounds, 3 parts by weight of the sodium salt of diisobutyl naphthalene-1-sulfonic acid, 10 parts by weight of the sodium salt of a lignosulfonic acid from a sulfite waste liquor and 7 parts by weight of pulverulent 45 silica gel; a spray mixture is obtained by finely distributing the mixture in water;

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- VI. An intimate mixture of 3 parts by weight of the active compounds and 97 parts by weight of finely divided kaolin; this dust comprises 3% by weight of active compound;
- 5 VII. An intimate mixture of 30 parts by weight of the active compounds, 92 parts by weight of pulverulent silica gel and 8 parts by weight of paraffin oil which had been sprayed onto the surface of this silica gel; this formulation imparts good adhesion to the active compound;
- 10 VIII. A stable aqueous dispersion of 40 parts by weight of the active compounds, 10 parts by weight of the sodium salt of a phenolsulfonic acid/urea/formaldehyde condensate, 2 parts by weight of silica gel and 48 parts by weight of water; this dispersion may be diluted further;
- 15 IX. A stable oily dispersion of 20 parts by weight of the active compounds, 2 parts by weight of the calcium salt of dodecylbenzenesulfonic acid, 8 parts by weight of fatty alcohol polyglycol ether, 20 parts by weight of the sodium salt of a phenolsulfonic acid/urea/formaldehyde condensate and 88 parts by weight of a paraffinic mineral oil.

20

Use Example

The synergistic activity of the mixtures according to the invention can be demonstrated by the following experiments:

25

The active compounds, separately or together, are formulated as a 10% emulsion in a mixture of 63% by weight of cyclohexanone and 27% by weight of emulsifier, and diluted with water to the desired concentration.

30

Evaluation is carried out by determining the infected leaf areas in percent. These percentages are converted into efficacies. The efficacy (\bar{W}) is calculated as follows using Abbot's formula:

35

$$W = (1 - \alpha/\beta) \cdot 100$$

α corresponds to the fungal infection of the treated plants in % and

40 β corresponds to the fungal infection of the untreated (control) plants in %

An efficacy of 0 means that the infection level of the treated plants corresponds to that of the untreated control plants; an efficacy of 100 means that the treated plants were not infected.

45

The expected efficacies of the mixtures of the active compounds were determined using Colby's formula [R.S. Colby, Weeds 15,

20-22 (1967)] and compared with the observed efficacies.

Colby's formula: $E = x + y - x \cdot y / 100$

- 5 E expected efficacy, expressed in % of the untreated control, when using the mixture of the active compounds A and B at the concentrations a and b
- x efficacy, expressed in % of the untreated control, when using active compound A at a concentration of a
- 10 y efficacy, expressed in % of the untreated control, when using active compound B at a concentration of b.

Use Example 1 - Activity against mildew of wheat

- 15 Leaves of potted wheat seedlings cv. "Kanzler" were sprayed to runoff point with an aqueous preparation of active compound which was prepared from a stock solution comprising 10% of active compound, 63% of cyclohexanone and 27% of emulsifier and, 24 h after the spray coating had dried on, dusted with spores of
- 20 mildew of wheat (*Erysiphe graminis forma specialis tritici*). The test plants were subsequently placed in climatized chambers at 20-24°C and 60-90% relative atmospheric humidity for 7 days. The extent of the development of the infection on the leaves was then determined visually.
- 25 The visually determined values for the percentage of infected leaf areas were converted into efficacies as % of the untreated control. An efficacy of 0 means the same degree of infection as in the untreated control, an efficacy of 100 means 0% infection.
- 30 The expected efficacies for active compound combinations were determined using Colby's formula (Colby, S.R. "Calculating synergistic and antagonistic responses of herbicide combinations", Weeds, 15 (1967), 20-22) and compared with the observed efficacies.
- 35 The components II used were the compounds II.79 and II.138 from Table 2.

The results of the tests are shown in Tables 1 and 2 below:

40

45

Table 1:

5	Ex.	Active compound	Conc. in ppm	Efficacy in % of the untreated control
	1C	without	(67% infected)	0
	2C	Compound II.79	1	55
			0.25	55
10	3C	Compound II.138	0.6	65
	4C	Compound I.a (common name: fenpropimorph)	0.25	55
	5C	Compound I.b (common name: fenpropidin)	0.25	55
15	6C	Compound I.c (common name: tridemorph)	1	0
			0.25	0

20
Table 2:

	Ex.	Mixture according to the invention (conc. in ppm)	Observed efficacy	Calculated efficacy*
25	7	0.25 ppm Ia + 0.25 ppm II.79	96	80
	8	1 ppm Ic + 1 ppm II.79	85	55
30	9	0.25 ppm Ic + 0.25 ppm II.79	90	55
	10	0.25 ppm Ib + 0.25 ppm II.79	93	80
	11	0.25 ppm Ia + 0.06 ppm II.138	100	84
35	12	0.25 ppm Ic + 0.06 ppm II.138	96	65
	13	0.25 ppm Ib + 0.06 ppm II.138	25	84

40 * Calculated using Colby's formula

The test results show that, for all mixing ratios, the observed efficacy is higher than the efficacy which had been calculated
45 beforehand using Colby's formula.

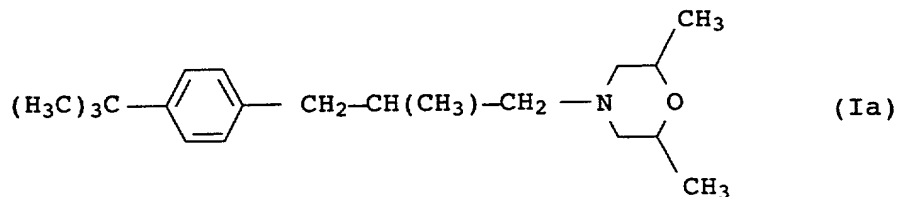
We claim:

1. A fungicidal mixture, comprising as active components

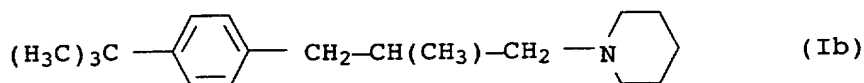
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a) a morpholine or piperidine derivative I selected from the group of the compounds Ia, Ib, Ic and Id

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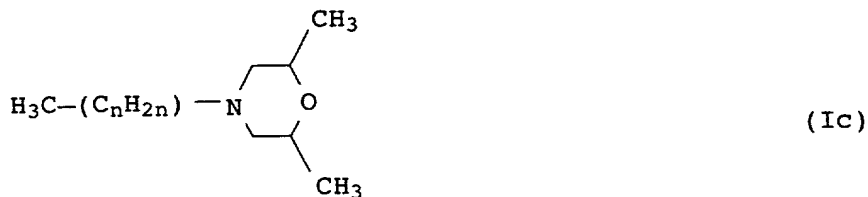


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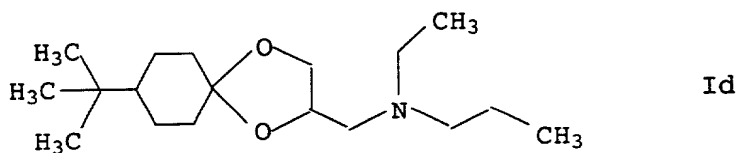
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[n = 10, 11, 12 (60 - 70%) or 13]

30



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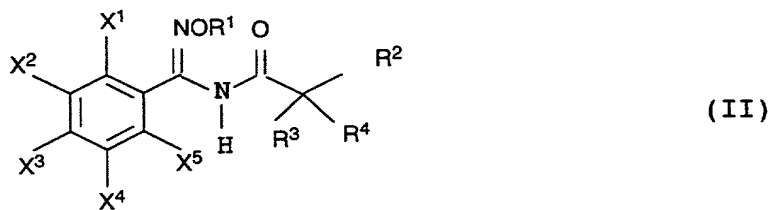
and

b) compounds of the formula II

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where the substituents X^1 to X^5 and R^1 to R^4 are as defined below:

15

X^1 is C_1 - C_4 -haloalkyl, C_1 - C_4 -haloalkoxy or halogen;

X^2 to X^5 are, independently of one another, hydrogen, halogen, C_1 - C_4 -alkyl, C_1 - C_4 -haloalkyl, C_1 - C_4 -alkoxy or C_1 - C_4 -haloalkoxy,

20

R^1 is C_1 - C_4 -alkyl, C_2 - C_6 -alkenyl, C_2 - C_6 -alkynyl, C_1 - C_4 -alkyl- C_3 - C_7 -cycloalkyl, where these radicals may carry substituents selected from the group consisting of halogen, cyano and C_1 - C_4 -alkoxy,

25

R^2 is a phenyl radical or a 5- or 6-membered saturated or unsaturated heterocyclyl radical having at least one heteroatom selected from the group consisting of N, O and S, where the cyclic radicals may have one to three substituents selected from the group consisting of halogen, C_1 - C_4 -alkyl, C_1 - C_4 -alkoxy, C_1 - C_4 -haloalkyl, C_1 - C_4 -haloalkoxy, C_1 - C_4 -alkoxy- C_2 - C_4 -alkenyl, C_1 - C_4 -alkoxy- C_2 - C_4 -alkynyl,

30

R^3 and R^4 are, independently of one another, hydrogen, C_1 - C_4 -alkyl, C_1 - C_4 -alkoxy, C_1 - C_4 -alkylthio, N- C_1 - C_4 -alkylamino, C_1 - C_4 -haloalkyl or C_1 - C_4 -haloalkoxy

35

in a synergistically effective amount.

40

2. A fungicidal mixture as claimed in claim 1, where in the compounds II, R^1 is C_1 - C_4 -alkyl or C_1 - C_4 -alkylene- C_3 - C_7 -cycloalkyl.

45

3. A fungicidal mixture as claimed in claim 1, where in the compounds II, R^2 is phenyl, thienyl, pyrazolyl, pyrrolyl, imidazolyl, thiazolyl, furyl, pyridazinyl or pyrimidinyl, and

these radicals may be substituted by halogen, C₁-C₄-alkoxy or C₁-C₄-alkyl.

4. A fungicidal mixture as claimed in claim 1, where in the
5 compounds II, R³ or R⁴ are hydrogen, fluorine, chlorine, methyl, ethyl, methoxy, thiomethyl or N-methyamino [sic].
5. A fungicidal mixture as claimed in claim 1, where in the
10 compounds II, X¹ is halo-C₁-C₆-alkyl, halo-C₁-C₆-alkoxy or halogen.
6. A fungicidal mixture as claimed in claim 1, where in the
compounds II, X² or X³ are hydrogen or halogen.
- 15 7. A fungicidal mixture as claimed in claim 1, where in the
compounds II, X⁴ is hydrogen, chlorine, fluorine, methoxy, ethoxy, trifluoromethyl or trifluoromethoxy.
8. A fungicidal mixture as claimed in claim 1, where in the
20 compounds II, X⁵ is hydrogen, chlorine, fluorine, methoxy, ethoxy, trifluoromethyl or trifluoromethoxy.
9. A fungicidal mixture as claimed in any one of the preceding
claims, which is conditioned in two parts, where one part
25 comprises one or more compounds I in a solid or liquid carrier and the other part comprises one or more compounds of the formula II in a solid or liquid carrier.
10. A method for controlling harmful fungi, which comprises
30 treating the fungi, their habitat or the materials, plants, seeds, soils, areas or spaces to be protected against fungal attack with a fungicidal mixture as claimed in any of claims 1 to 9, where the compounds I and one or more compounds of the formulae [sic] II can be applied simultaneously, that is
35 either together or separately, or successively.

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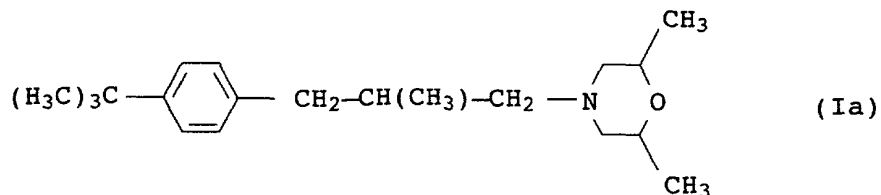
Abstract

Fungicidal mixtures, comprising as active components

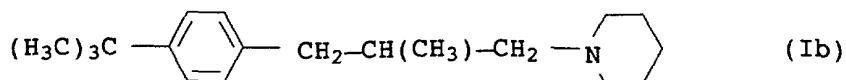
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- a) a morpholine or piperidine derivative I selected from the group of the compounds Ia, Ib, Ic and Id

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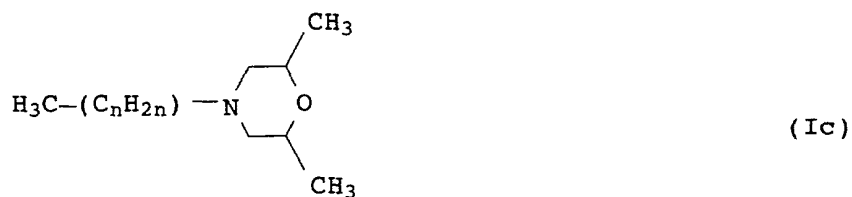


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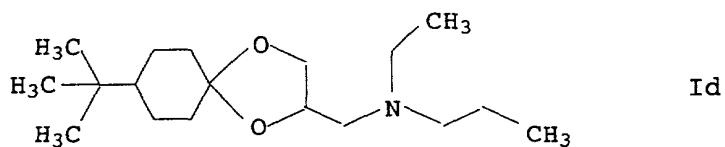
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[n = 10, 11, 12 (60 - 70%) or 13]

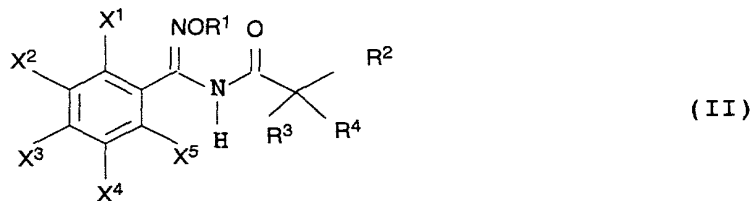
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and

- b) compounds of the formula II

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where the substituents X^1 to X^5 and R^1 to R^4 are as defined below:

X^1 is C_1 - C_4 -haloalkyl, C_1 - C_4 -haloalkoxy or halogen;

5 X^2 to X^5 are, independently of one another, hydrogen, halogen, C_1 - C_4 -alkyl, C_1 - C_4 -haloalkyl, C_1 - C_4 -alkoxy or C_1 - C_4 -haloalkoxy;

10 R^1 is C_1 - C_4 -alkyl, C_2 - C_6 -alkenyl, C_2 - C_6 -alkynyl, C_1 - C_4 -alkyl- C_3 - C_7 -cycloalkyl, C_1 - C_4 -alkyl- C_3 - C_7 -cycloalkenyl, where these radicals may carry substituents selected from the group consisting of halogen, cyano and C_1 - C_4 -alkoxy,

15 R^2 is a phenyl radical or a 5- or 6-membered saturated or unsaturated heterocyclyl radical having at least one heteroatom selected from the group consisting of N, O and S, where the cyclic radicals may have one to three substituents selected from the group consisting of halogen, C_1 - C_4 -alkyl, C_1 - C_4 -alkoxy, C_1 - C_4 -haloalkyl, C_1 - C_4 -haloalkoxy,
20 C_1 - C_4 -alkoxy- C_2 - C_4 -alkenyl, C_1 - C_4 -alkoxy- C_2 - C_4 -alkynyl,

R^3 and R^4 are, independently of one another, hydrogen, C_1 - C_4 -alkyl, C_1 - C_4 -alkoxy, C_1 - C_4 -alkylthio, N- C_1 - C_4 -alkylamino, C_1 - C_4 -haloalkyl or C_1 - C_4 -haloalkoxy

25 in a synergistically effective amount are described.

30

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Declaration, Power of Attorney

Page 1 of 5

0050/049651

We (I), the undersigned inventor(s), hereby declare(s) that:

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

Fungicidal mixtures based on morpholine or piperidine derivatives
and oxime ether derivatives

the specification of which

☒ is attached hereto.

[] was filed on _____ as

Application Serial No. _____

and amended on _____.

[x] was filed as PCT international application

Number PCT/EP99/09803

on December 11, 1999

and was amended under PCT Article 19

on _____ (if applicable).

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application(s)

Application No.	Country	Day/Month/Year	Priority Claimed
19858911.5	Germany	19 December 1998	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No

We (I) hereby claim the benefit under Title 35, United States Codes, § 119(e) of any United States provisional application(s) listed below.

(Application Number)

(Filing Date)

(Application Number)

(Filing Date)

We (I) hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

Application Serial No.	Filing Date	Status (pending, patented, abandoned)
_____	_____	_____
_____	_____	_____
_____	_____	_____
_____	_____	_____

And we (I) hereby appoint **Messrs. HERBERT. B. KEIL**, Registration Number 18,967; and **RUSSEL E. WEINKAUF**, Registration Number 18,495; the address of both being Messrs. Keil & Weinkauff, 1101 Connecticut Ave., N.W., Washington, D.C. 20036 (telephone 202-659-0100), our attorneys, with full power of substitution and revocation, to prosecute this application, to make alterations and amendments therein, to sign the drawings, to receive the patent, and to transact all business in the Patent Office connected therewith.

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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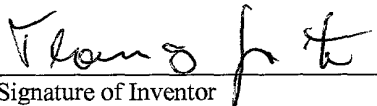
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Page 5 of 5

0050/049651

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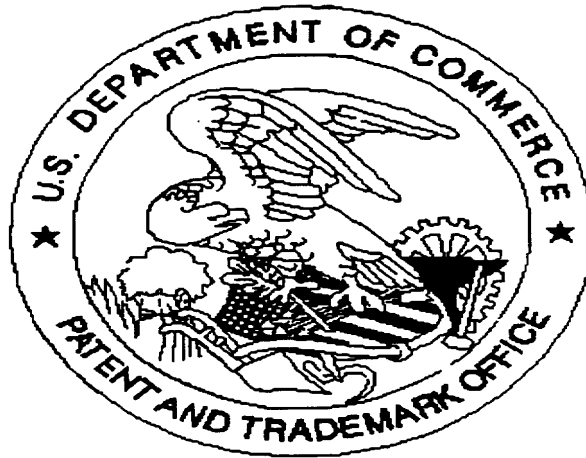

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